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Ottawa Hull K1A 0C9

(21) (A1) 2,106,497
(22) 1993/09/20
(43) 1994/03/22

(51) INTL.CL.[5] C07D-277/56; C07D-213/82; C07D-231/14; C07D-307/68; C07D-309/28; C07D-327/06; C07D-333/38; C07C-259/10; A01N-037/28; A01N-043/02; A01N-043/40; A01N-043/78

(19)(CA) **APPLICATION FOR CANADIAN PATENT** (12)

(54) N-Hydroxy-N-Phenylcarboxamides, Their Preparation and Compositions Containing Them for Controlling Harmful Fungi

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(30) (DE) P 42 31 518.2 1992/09/21

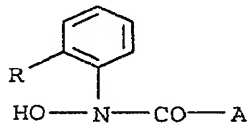
(57) 7 Claims

2106497

N-Hydroxy-N-phenylcarboxamides, their manufacture, and agents containing them for for combatting injurious fungi

ABSTRACT OF THE DISCLOSURE:

1. N-Hydroxy-N-phenylcarboxamides of the formula I

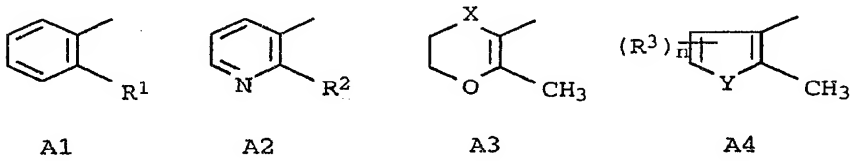


I

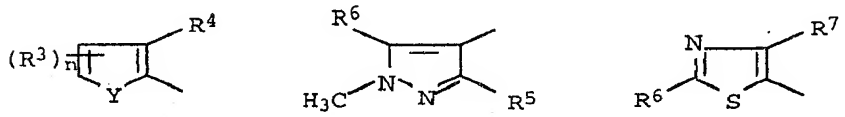
where:

R is substituted or unsubstituted alkyl, alkoxy, alkenyl, alkenyloxy, alkynyl, alkynyloxy, cycloalkyl, cycloalkenyl, cycloalkyloxy, cycloalkenyloxy or phenyl;

A is one of the radicals A1 to A7



A1 A2 A3 A4



A5 A6 A7

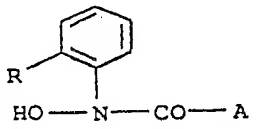
where

X is $-CH_2-$, $-S-$, $-SO-$ or $-SO_2-$;
Y is $-O-$ or $-S-$;
R^1, R^2, R^4, R^5 and R^7 are halogen, alkyl or haloalkyl;
R^3 and R^6 are hydrogen, halogen or alkyl;
n is 1 or 2;

methods of manufacturing them, agents containing them, and their use for combatting injurious fungi.

N-HYDROXY-N-PHENYLCARBOXAMIDES, THEIR PREPARATION AND COMPOSITIONS CONTAINING THEM FOR CONTROLLING HARMFUL FUNGI

The present invention relates to N-hydroxy-N-phenylcarboxamides of the formula I

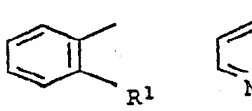


I

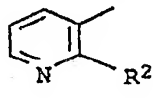
where the substituents have the following meanings:

R is C_2-C_{12}-alkyl, C_2-C_{12}-alkoxy, C_3-C_{12}-alkenyl, C_3-C_{12}-alkenyloxy, C_3-C_6-alkynyl, C_3-C_6-alkynyloxy, where these groups can be partially or completely halogenated; C_3-C_7-cycloalkyl, C_4-C_7-cycloalkenyl, C_3-C_7-cycloalkoxy or C_4-C_7-cycloalkenyloxy, where these rings can carry one to 3 C_1-C_4-alkyls; phenyl which can carry one to five halogen atoms and/or one to three of the following radicals: C_1-C_4-alkyl, C_1-C_4-haloalkyl, C_1-C_4-alkoxy, C_1-C_4-haloalkoxy, C_1-C_4-alkylthio or C_1-C_4-haloalkylthio;

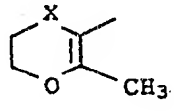
A is a cyclic radical from the group consisting of the formulae A1 to A7:



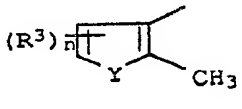
A1



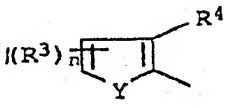
A2



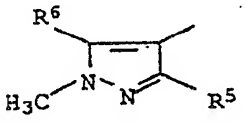
A3



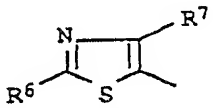
A4



A5



A6



A7

where the substituents have the following meanings:

X is $-CH_2-$, $-S-$, $-SO-$ or $-SO_2-$;

Y is $-O-$ or $-S-$;

R^1, R^2, R^4, R^5 and R^7 are halogen, C_1-C_4-alkyl or

C_1-C_4-haloalkyl;

R^3 and R^6 are hydrogen, halogen or C_1-C_4-alkyl;

n is 1 or 2, where the radicals R^3 can be different if the value of n is 2.

The invention additionally relates to the preparation of these compounds, compositions containing them and their use for controlling harmful fungi, in particular Botrytis.

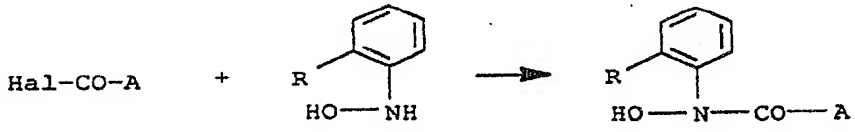
N-(2-Chlorophenyl)-2-chloronicotinamide is known from the literature as a fungicidal active compound (DE-A 2 417 216).

It is an object of the present invention to provide novel fungicidally active compounds having an improved spectrum of action.

We have found that this object is achieved by the compounds I defined at the beginning.

We have additionally found processes for preparing these compounds, compositions containing them and their use for controlling harmful fungi.

The compounds I are in general obtained by reacting a carboxylic acid halide of the formula II in a manner known per se (eg. J. March, Advanced Organic Chemistry, 2nd Ed., 1977, 382 ff., McGraw-Hill) with an N-hydroxyaniline of the formula III in the presence of a base.



II III I

The radical Hal in the formula II is a halogen such as chlorine, bromine or iodine, in particular chlorine or bromine.

This reaction is customarily carried out at temperatures from -20°C to 100°C, preferably -10°C to

50°C.

Suitable solvents are:
aliphatic hydrocarbons such as pentane, hexane, cyclohexane and petroleum ether, aromatic hydrocarbons such as toluene, o-, m- and p-xylene, halogenated hydrocarbons such as dichloromethane, chloroform and chlorobenzene, ethers such as diethyl ether, diisopropyl ether, tert-butyl methyl ether, dioxane, anisole and tetrahydrofuran, nitriles such as acetonitrile and propionitrile, ketones such as acetone, methyl ethyl ketone, diethyl ketone and tert-butyl methyl ketone, alcohols such as methanol, ethanol, n-propanol, isopropanol, n-butanol and tert-butanol, and also dimethyl sulfoxide and dimethylformamide, particularly preferably toluene, xylene and tetrahydrofuran.

Mixtures of the solvents mentioned can also be used.

Suitable bases are generally inorganic compounds such as alkali metal and alkaline earth metal hydroxides such as lithium hydroxide, sodium hydroxide, potassium hydroxide and calcium hydroxide, alkali metal and alkaline earth metal oxides such as lithium oxide, sodium oxide, calcium oxide and magnesium oxide, alkali metal and alkaline earth metal hydrides such as lithium hydride, sodium hydride, potassium hydride and calcium hydride, alkali metal amides such as lithium amide, sodium amide and potassium amide, alkali metal and alkaline earth metal carbonates such as lithium carbonate and calcium carbonate and also alkali metal hydrogencarbonates such as sodium hydrogencarbonate, and organometallic compounds, in particular alkali metal alkyls such as methyllithium, butyllithium and phenyllithium, alkylmagnesium halides such as methylmagnesium chloride and also alkali metal and alkaline earth metal alkoxides such as sodium methoxide, sodium ethoxide, potassium ethoxide, potassium tert-butoxide and dimethoxymagnesium, additionally organic bases, eg. tertiary amines such as

trimethylamine, triethylamine, tri-isopropylethylamine and N-methylpiperidine, pyridine, substituted pyridines such as collidine, lutidine and 4-dimethylaminopyridine and also bicyclic amines.

Sodium hydrogencarbonate, sodium carbonate, triethylamine and pyridine are particularly preferred.

The bases are in general employed in equimolar amounts based on the compound II. However, they can also be used in an excess of from 5 mol% to 30 mol%, preferably 5 mol% to 10 mol%, or - in the case of the use of tertiary amines - if appropriate as a solvent.

The starting materials are in general reacted with one another in equimolar amounts. It may be advantageous for the yield to employ II in an excess of from 1 mol% to 20 mol%, preferably 1 mol% to 10 mol%, based on III.

The starting substances of the formulae II and III needed for preparing the compounds I are known in the literature (Houben-Weyl, Methoden der org. Chemie (Methods of Organic Chemistry), Vol. 10/1, pp. 1138-1148) or can be prepared according to the literature cited.

With respect to their use in fungicidal compositions, suitable compounds of the formula I are those in which the substituents have the following meanings:

R is C_2-C_{12}-alkyl such as ethyl and straight-chain or branched propyl, butyl, pentyl, hexyl, heptyl, octyl, nonyl, decyl, undecyl and dodecyl, particularly straight-chain or branched C_3-C_{10}-alkyl such as propyl, 1-methylethyl, butyl, 1-methylpropyl, 2-methylpropyl, 1,1-dimethylethyl, n-pentyl, 1-methylbutyl, 2-methylbutyl, 3-methylbutyl, 1,2-dimethylpropyl, 1,1-dimethylpropyl, 2,2-dimethylpropyl, 1-ethylpropyl, n-hexyl, 1-methylpentyl, 2-methylpentyl, 3-methylpentyl, 4-methylpentyl, 1,2-dimethylbutyl, 1,3-dimethylbutyl, 1,3-dimethylbutyl, 2,3-dimethylbutyl, 1,1-dimethylbutyl, 2,2-dimethylbutyl, 3,3-dimethyl-

butyl, 1,1,2-trimethylpropyl, 1,2,2-trimethylpropyl, 1-ethylbutyl, 2-ethylbutyl, 1-ethyl-3-methylpropyl, n-heptyl, 1-methylhexyl, 1-ethylpentyl, 2-ethylpentyl, 1-propylbutyl, octyl, 1-methylheptyl, 2-methylheptyl, 1-ethylhexyl, 2-ethylhexyl, 1-propylpentyl, 2-propylpentyl, nonyl, 1-methyloctyl, 2-methyloctyl, 1-ethylheptyl, 2-ethylheptyl, 1-propylhexyl, 2-propylhexyl, decyl, 1-methylnonyl, 2-methylnonyl, 1-ethyloctyl, 2-ethyloctyl, 1-propylheptyl and 2-propylheptyl, in particular propyl, 1-methylethyl, butyl, 1-methylbutyl, 2-methylbutyl, 1,1-dimethylethyl, pentyl, 1-methylbutyl, hexyl, heptyl and 1-methylheptyl, where these groups can be partially or completely halogenated, ie. the hydrogens of these groups can be partially or completely replaced by halogens such as fluorine, chlorine and bromine, in particular fluorine and chlorine, for example haloalkyl such as chloromethyl, dichloromethyl, trichloromethyl, fluoromethyl, difluoromethyl, trifluoromethyl, chlorofluoromethyl, dichlorofluoromethyl, chlorodifluoromethyl, 1-fluoroethyl, 2-fluoroethyl, 2,2-difluoroethyl, 2,2,2-trifluoroethyl, 2-chloro-2-fluoroethyl, 2-chloro-2,2-difluoroethyl, 2,2-dichloro-2-fluoroethyl, 2,2,2-trichloroethyl and pentafluoroethyl;

C_2-C_{12}-alkoxy such as ethoxy and straight-chain or branched propoxy, butoxy, pentoxy, hexyloxy, heptyloxy, octyloxy, nonyloxy, decyloxy, undecyloxy and dodecyloxy, particularly straight-chain or branched C_2-C_{10}-alkoxy such as ethoxy, propoxy, 1-methylethoxy, butoxy, 1-methylpropoxy, 2-methylpropoxy, 1,1-dimethylethoxy, n-pentoxy, 1-methylbutoxy, 2-methylbutoxy, 3-methylbutoxy, 1,2-dimethylpropoxy, 1-ethylpropoxy, n-hexyloxy, 1-methylpentoxy, 2-methylpentoxy, 3-methylpentoxy, 4-methylpentoxy, 1,2-dimethylbutoxy, 1,3-dimethylbutoxy,

2,3-dimethylbutoxy, 1,2-dimethylbutoxy, 2,2-dimethylbutoxy, 3,3-dimethylbutoxy, 1,1,2-trimethylpropoxy, 1,2,2-trimethylpropoxy, 1-ethylbutoxy, 2-ethylbutoxy, 1-ethyl-2-methylpropoxy, n-heptyloxy, 1-methylhexyloxy, 2-methylhexyloxy, 3-methylhexyloxy, 4-methylhexyloxy, 5-methylhexyloxy, 1-ethylpentoxy, 2-ethylpentoxy, 1-propylbutoxy, octyloxy, 1-methylheptyloxy, 2-methylheptyloxy, 1-ethylhexyloxy, 2-ethylhexyloxy, 1-propylpentoxy, 2-propylpentoxy, nonyloxy, 1-methyloctyloxy, 2-methyloctyloxy, 1-ethylheptyloxy, 2-ethylheptyloxy, 1-propylhexyloxy, 2-propylhexyloxy, decyloxy, 1-methylnonyloxy, 2-methylnonyloxy, 1-ethyloctyloxy, 2-ethyloctyloxy, 1-propylheptyloxy and 2-propylheptyloxy, in particular ethoxy, propoxy, 1-methylethoxy, butoxy, 1-methylpropoxy, 2-methylpropoxy, 1,1-dimethylethoxy, pentoxy, hexyloxy and 2-ethylhexyloxy, where these groups can be partially or completely halogenated, ie. the hydrogens of these groups can be partially or completely replaced by halogens such as fluorine, chlorine and bromine, in particular fluorine and chlorine, for example haloalkoxy such as chloromethoxy, dichloromethoxy, trichloromethoxy, fluoromethoxy, difluoromethoxy, trifluoromethoxy, chlorofluoromethoxy, dichlorofluoromethoxy, chlorodifluoromethoxy, 1-fluoroethoxy, 2-fluoroethoxy, 2,2-difluoroethoxy, 2,2,2-trifluoroethoxy, 2-chloro-2-fluoroethoxy, 2-chloro-2,2-difluoroethoxy, 2,2-dichloro-2-fluoroethoxy, 2,2,2-trichloroethoxy and pentafluoroethoxy;

C_3-C_{12}-alkenyl such as straight-chain or branched propenyl, butenyl, pentenyl, hexenyl, heptenyl, octenyl, nonenyl, decenyl, undecenyl and dodecenyl, particularly straight-chain or branched C_3-C_{10}-alkenyl such as 2-propenyl, 2-butenyl, 3-butenyl, 1-methyl-2-propenyl, 2-methyl-2-propenyl,

2-pentenyl, 3-pentenyl, 4-pentenyl, 1-methyl-2-butenyl, 2-methyl-2-butenyl, 3-methyl-2-butenyl, 1-methyl-3-butenyl, 2-methyl-3-butenyl, 3-methyl-3-butenyl, 1,1-dimethyl-2-propenyl, 1,2-dimethyl-2-propenyl, 1-ethyl-2-propenyl, 2-hexenyl, 3-hexenyl, 4-hexenyl, 5-hexenyl, 1-methyl-2-pentenyl, 2-methyl-2-pentenyl, 3-methyl-2-pentenyl, 4-methyl-2-pentenyl, 1-methyl-3-pentenyl, 2-methyl-3-pentenyl, 3-methyl-3-pentenyl, 4-methyl-3-pentenyl, 1-methyl-4-pentenyl, 2-methyl-4-pentenyl, 3-methyl-4-pentenyl, 4-methyl-4-pentenyl, 1,1-dimethyl-2-butenyl, 1,1-dimethyl-3-butenyl, 1,2-dimethyl-2-butenyl, 1,2-dimethyl-3-butenyl, 1,3-dimethyl-2-butenyl, 1,3-dimethyl-3-butenyl, 2,2-dimethyl-3-butenyl, 2,3-dimethyl-2-butenyl, 2,3-dimethyl-3-butenyl, 1-ethyl-2-butenyl, 1-ethyl-3-butenyl, 2-ethyl-2-butenyl, 2-ethyl-3-butenyl, 1,1,2-trimethyl-2-propenyl, 1-ethyl-1-methyl-2-propenyl, 1-ethyl-2-methyl-2-propenyl, 1-methyl-2-pentenyl, 2-methyl-2-pentenyl, 1-methyl-3-pentenyl, 2-methyl-3-pentenyl, 1-methyl-2-hexenyl, 2-methyl-2-hexenyl, 1-methyl-3-hexenyl, 2-methyl-3-hexenyl, 1-ethyl-2-pentenyl, 2-ethyl-2-pentenyl, 1-ethyl-3-pentenyl, 2-ethyl-3-pentenyl, 1-methyl-2-heptenyl, 2-methyl-2-heptenyl, 1-methyl-3-heptenyl, 2-methyl-3-heptenyl, 1-ethyl-2-hexenyl, 2-ethyl-2-hexenyl, 1-ethyl-3-hexenyl, 2-ethyl-3-hexenyl, 1-methyl-2-octenyl, 2-methyl-2-octenyl, 1-methyl-3-octenyl, 2-methyl-3-octenyl, 1-ethyl-2-heptenyl, 2-ethyl-2-heptenyl, 1-ethyl-3-heptenyl, 2-ethyl-3-heptenyl, 1-ethyl-2-octenyl, 2-ethyl-2-octenyl, 1-ethyl-3-octenyl and 2-ethyl-3-octenyl, in particular 1-propenyl, 2-propenyl, 1-methylethenyl, 1-methyl-2-propenyl, 2-methyl-2-propenyl, 1-ethyl-2-propenyl, 1-methyl-2-butenyl, 1-ethyl-2-butenyl, 1-(1-methylethyl)-2-butenyl, 1-butyl-2-butenyl, 1-methyl-

2-pentenyl and 1,4-dimethyl-2-pentenyl, where these groups can be partially or completely halogenated, ie. the hydrogens of these groups can be partially or completely replaced by halogens such as fluorine, chlorine and bromine, in particular fluorine and chlorine, in particular 3-chloro-2-propenyl and 2,3-dichloro-2-propenyl;

C_3-C_{12}-alkenyloxy such as straight-chain or branched propenyloxy, butenyloxy, pentenyloxy, hexenyloxy, heptenyloxy, octenyloxy, nonenyloxy, decenyloxy, undecenyloxy and dodecenyloxy, particularly straight-chain or branched C_3-C_{10}-alkenyloxy such as 2-propenyloxy, 2-butenyloxy, 3-butenyloxy, 1-methyl-2-propenyloxy, 2-methyl-2-propenyloxy, 2-pentenyloxy, 3-pentenyloxy, 4-pentenyloxy, 1-methyl-2-butenyloxy, 2-methyl-2-butenyloxy, 3-methyl-2-butenyloxy, 1-methyl-3-butenyloxy, 2-methyl-3-butenyloxy, 3-methyl-3-butenyloxy, 1,1-dimethyl-2-propenyloxy, 1,2-dimethyl-2-propenyloxy, 1-ethyl-2-propenyloxy, 2-hexenyloxy, 3-hexenyloxy, 4-hexenyloxy, 5-hexenyloxy, 1-methyl-2-pentenyloxy, 2-methyl-2-pentenyloxy, 3-methyl-2-pentenyloxy, 4-methyl-2-pentenyloxy, 1-methyl-3-pentenyloxy, 2-methyl-3-pentenyloxy, 3-methyl-3-pentenyloxy, 4-methyl-3-pentenyloxy, 1-methyl-4-pentenyloxy, 2-methyl-4-pentenyloxy, 3-methyl-4-pentenyloxy, 4-methyl-4-pentenyloxy, 1,1-dimethyl-2-butenyloxy, 1,1-dimethyl-3-butenyloxy, 1,2-dimethyl-2-butenyloxy, 1,2-dimethyl-3-butenyloxy, 1,3-dimethyl-2-butenyloxy, 1,3-dimethyl-3-butenyloxy, 2,2-dimethyl-3-butenyloxy, 2,3-dimethyl-2-butenyloxy, 2,3-dimethyl-3-butenyloxy, 1-ethyl-2-butenyloxy, 1-ethyl-3-butenyloxy, 2-ethyl-2-butenyloxy, 2-ethyl-3-butenyloxy, 1,1,2-trimethyl-2-propenyloxy, 1-ethyl-1-methyl-2-propenyloxy, 1-ethyl-2-methyl-2-propenyloxy, 1-methyl-2-pentenyl-

oxy, 2-methyl-2-pentenyloxy, 1-methyl-3-pentenyloxy, 2-methyl-3-pentenyloxy, 1-methyl-2-hexenyloxy, 2-methyl-2-hexenyloxy, 1-methyl-3-hexenyloxy, 2-methyl-3-hexenyloxy, 1-ethyl-2-pentenyloxy, 2-ethyl-2-pentenyloxy, 1-ethyl-3-pentenyloxy, 2-ethyl-3-pentenyloxy, 1-methyl-2-heptenyloxy, 2-methyl-2-heptenyloxy, 1-methyl-3-heptenyloxy, 2-methyl-3-heptenyloxy, 1-ethyl-2-hexenyloxy, 2-ethyl-2-hexenyloxy, 1-ethyl-3-hexenyloxy, 2-ethyl-3-hexenyloxy, 1-methyl-2-octenyloxy, 2-methyl-2-octenyloxy, 1-methyl-3-octenyloxy, 2-methyl-3-octenyloxy, 1-ethyl-2-heptenyloxy, 2-ethyl-2-heptenyloxy, 1-ethyl-3-heptenyloxy, 2-ethyl-3-heptenyloxy, 1-ethyl-2-octenyloxy, 2-ethyl-2-octenyloxy, 1-ethyl-3-octenyloxy and 2-ethyl-3-octenyloxy, in particular 2-propenyloxy, 1-methyl-2-propenyloxy, 2-methyl-2-propenyloxy, 2-pentenyloxy, 3-pentenyloxy, 1-methyl-2-butenyloxy and 1-methyl-2-pentenyloxy, where these groups can be partially or completely halogenated, ie. the hydrogens of these groups can be partially or completely replaced by halogens such as fluorine, chlorine and bromine, in particular fluorine and chlorine, in particular 3-chloro-2-propenyloxy, 2,3-dichloro-2-propenyloxy and 2,3,3-trichloro-2-propenyloxy;

C_3-C_6-alkynyl such as 2-propynyl, 2-butynyl, 3-butynyl, 1-methyl-2-propynyl, 2-pentynyl, 3-pentynyl, 4-pentynyl, 1-methyl-3-butynyl, 2-methyl-3-butynyl, 1-methyl-2-butynyl, 1,1-dimethyl-2-propynyl, 1-ethyl-2-propynyl, 2-hexynyl, 3-hexynyl, 4-alkynyl, 5-hexynyl, 1-methyl-2-pentynyl, 1-methyl-3-pentynyl, 1-methyl-4-pentynyl, 2-methyl-3-pentynyl, 2-methyl-4-pentynyl, 3-methyl-4-pentynyl, 4-methyl-2-pentynyl, 1,2-dimethyl-2-butynyl, 1,1-dimethyl-3-butynyl, 1,2-dimethyl-3-butynyl, 2,2-dimethyl-3-

butynyl, 1-ethyl-2-butynyl, 1-ethyl-3-butynyl, 2-ethyl-3-butynyl and 1-ethyl-1-methyl-2-propynyl, in particular 2-propynyl, 2-butynyl and 3-butynyl, where these groups can be partially or completely halogenated, ie. the hydrogens of these groups can be partially or completely replaced by halogens such as fluorine, chlorine and bromine, in particular fluorine and chlorine, for example 3-chloro-2-propynyl, 3-chloro-2-butynyl and 4-chloro-3-butynyl;

C_3-C_6-alkynyloxy such as 2-propynyloxy, 2-butynyloxy, 3-butynyloxy, 1-methyl-2-propynyloxy, 2-pentynyloxy, 3-pentynyloxy, 3-pentynyloxy, 4-pentynyloxy, 1-methyl-3-butynyloxy, 2-methyl-3-butynyloxy, 1-methyl-2-butynyloxy, 1,1-dimethyl-2-propionyloxy, 1-ethyl-2-propynyloxy, 2-hexynyloxy, 3-hexynyloxy, 4-alkynyloxy, 5-hexynyloxy, 1-methyl-2-pentynyloxy, 1-methyl-3-pentynyloxy, 1-methyl-4-pentynyloxy, 2-methyl-3-pentynyloxy, 2-methyl-4-pentynyloxy, 3-methyl-4-pentynyloxy, 4-methyl-3-pentynyloxy, 1,1-dimethyl-2-butynyloxy, 1,1-dimethyl-3-butynyloxy, 1,2-dimethyl-3-butynyloxy, 2,2-dimethyl-3-butynyloxy, 1-ethyl-2-butynyloxy, 1-ethyl-3-butynyloxy, 2-ethyl-3-butynyloxy and 1-ethyl-1-methyl-2-propynyloxy, preferably 2-propynyloxy, 2-butynyloxy, 1-methyl-2-propynyloxy and 1-methyl-2-butynyloxy, 2-propynyloxy, 2-butynyloxy, 3-butynyloxy and 1-methyl-2-propynyloxy, where these groups can be partially or completely halogenated, ie. the hydrogens of these groups can be partially or completely replaced by halogens such as fluorine, chlorine and bromine, in particular fluorine and chlorine, for example 3-chloro-2-propynyloxy, 3-chloro-2-butynyloxy and 4-chloro-3-butynyloxy;

C_3-C_7-cycloalkyl such as cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl and cycloheptyl, where these

rings can carry one to 3 C_1-C_4-alkyls such as methyl, ethyl, propyl, 1-methylethyl, butyl, 1-methylpropyl, 2-methylpropyl and 1,1-dimethylethyl;

C_4-C_7-cycloalkenyl such as cyclobutenyl, cyclopentenyl, cyclohexenyl and cycloheptenyl, where these rings can carry one to three C_1-C_4-alkyls such as methyl, ethyl, propyl, 1-methylethyl, butyl, 1-methylpropyl, 2-methylpropyl and 1,1-dimethylethyl;

C_3-C_7-cycloalkoxy such as cyclopropoxy, cyclobutoxy, cyclopentoxy, cyclohexyloxy and cycloheptyloxy, where these rings can carry one to 3 C_1-C_4-alkyls such as methyl, ethyl, propyl, 1-methylethyl, butyl, 1-methylpropyl, 2-methylpropyl and 1,1-dimethylethyl;

or C_4-C_7-cycloalkenyloxy such as 1-cyclobutenyloxy, 2-cyclobutenyloxy, 1-cyclopentenyloxy, 2-cyclopentenyloxy, 3-cyclopentenyloxy, 1-cyclohexenyloxy, 2-cyclohexenyloxy, 3-cyclohexenyloxy, 1-cycloheptenyloxy, 2-cycloheptenyloxy, 3-cycloheptenyloxy and 4-cycloheptenyloxy, where these rings can carry one to 3 C_1-C_4-alkyls such as methyl, ethyl, propyl, 1-methylethyl, butyl, 1-methylpropyl, 2-methylpropyl and 1,1-dimethylethyl;

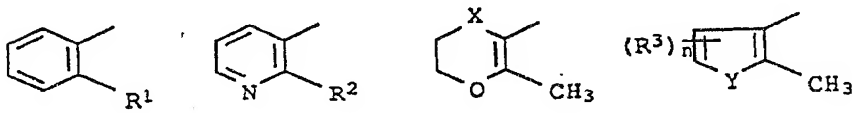
phenyl, which can carry one to five halogens such as fluorine, chlorine, bromine and iodine, in particular fluorine, chlorine and bromine, and/or one to three of the following radicals:

- C_1-C_4-alkyl as mentioned above;
- C_1-C_4-haloalkyl as mentioned above;
- C_1-C_4-alkoxy as mentioned above;
- C_1-C_4-haloalkoxy as mentioned above;
- C_1-C_4-alkylthio such as methylthio, ethylthio,

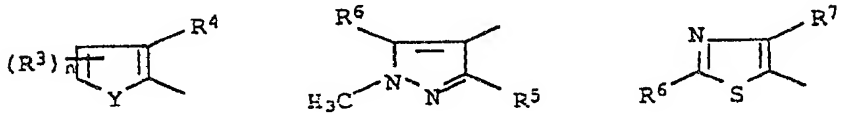
propylthio, 1-methylethylthio, butylthio, 1-methylpropylthio, 2-methylpropylthio and 1,1-dimethylethylthio;

- or C_1-C_4-haloalkylthio, particularly C_1-C_2-haloalkylthio such as chloromethylthio, dichloromethylthio, trichloromethylthio, fluoromethylthio, difluoromethylthio, trifluoromethylthio, chlorofluoromethylthio, dichlorofluoromethylthio, chlorodifluoromethylthio, 1-fluoroethylthio, 2-fluoroethylthio, 2,2-difluoroethylthio, 2,2,2-trifluoroethylthio, 2-chloro-2-fluoroethylthio, 2-chloro-2,2-difluoroethylthio, 2,2-dichloro-2-fluoroethylthio, 2,2,2-trichloroethylthio and pentafluoroethylthio.

A is a cyclic radical from the group consisting of the formulae A1 to A7:



A1 A2 A3 A4



A5 A6 A7

where the substituents have the following meanings:

X is $-CH_2-$, $-S-$, $-SO-$ or $-SO_2-$;

Y is $-O-$ or $-S-$;

R^1, R^2, R^4, R^5 and R^7 independently of one another are halogen such as fluorine, chlorine and bromine, C_1-C_4-alkyl as mentioned above, or C_1-C_4-haloalkyl as mentioned above;

R^3 and R^6 independently of one another are hydrogen, halogen such as fluorine, chlorine and bromine or

C_1-C_4-alkyl as mentioned above;

n is 1 or 2, where the radicals R^3 can be different if the value of n is 2.

With respect to the biological action, particularly preferred compounds of the formula I are those in which R has the abovementioned meanings and A is a cyclic radical from the group consisting of the formulae A1 to A7, where X and Y have the abovementioned meaning and the substituents are the following radicals:

R^1 is halogen such as fluorine, chlorine and bromine, methyl or C_1-haloalkyl such as chloromethyl, dichloromethyl, trichloromethyl, fluoromethyl, difluoromethyl, trifluoromethyl, chlorofluoromethyl, dichlorofluoromethyl and chlorodifluoromethyl;

R^2 is halogen such as fluorine, chlorine and bromine or C_1-haloalkyl such as chloromethyl, dichloromethyl, trichloromethyl, fluoromethyl, difluoromethyl, trifluoromethyl, chlorofluoromethyl, dichlorofluoromethyl and chlorodifluoromethyl;

R^3 is hydrogen or methyl;

n is 1 or 2, where the radicals R^3 can be different if the value of n is 2;

R^4 is halogen such as fluorine, chlorine and bromine or methyl;

R^5 is methyl or C_1-haloalkyl such as chloromethyl, dichloromethyl, trichloromethyl, fluoromethyl, difluoromethyl, trifluoromethyl, chlorofluoromethyl, dichlorofluoromethyl and chlorodifluoromethyl;

R^6 is hydrogen, halogen such as fluorine, chlorine and bromine or methyl;

R^7 is halogen such as fluorine, chlorine and bromine, methyl or C_1-haloalkyl such as chloromethyl, dichloromethyl, trichloromethyl, fluoromethyl, difluoromethyl, trifluoromethyl, chlorofluoromethyl, dichlorofluoromethyl and chlorodifluoromethyl.

In particular, those compounds of the formula I are preferred in which R has the abovementioned meaning

and A is a cyclic radical from the group consisting of the formulae A1 to A7, where X and Y have the above-mentioned meaning and the substituents are the following groups:

R^1 is chlorine, bromine, iodine, methyl or trifluoromethyl;

R^2 is chlorine or trifluoromethyl;

R^3 is hydrogen or methyl;

n is 1 or 2, where the radicals R^3 can be different if the value of n is 2;

R^4 is chlorine or methyl;

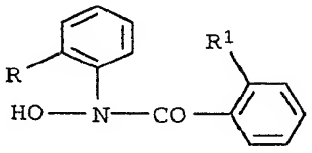
R^5 is methyl, difluoromethyl or trifluoromethyl;

R^6 is hydrogen, chlorine or methyl;

R^7 is chlorine, methyl or trifluoromethyl.

Particularly preferred compounds of the formula I are summarized in the following Tables A to G.

Table A



I.1

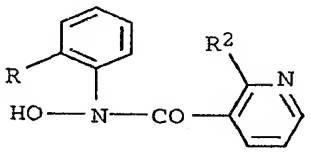
R^1	R
CF_3	i-C_3H_7
CF_3	n-C_3H_7
CF_3	n-C_4H_9
CF_3	sec.-C_4H_9
CF_3	i-C_4H_9
CF_3	tert.-C_4H_9
CF_3	n-C_5H_{11}
CF_3	sec-C_5H_{11}
CF_3	n-C_6H_{13}
CF_3	n-C_7H_{15}
CF_3	sec.-C_7H_{15}
CF_3	1-methylvinyl
CF_3	2-methylvinyl
CF_3	allyl
CF_3	2-methylallyl
CF_3	2-ethylallyl
CF_3	1-methylallyl
CF_3	1-ethylallyl
CF_3	1-methyl-2-butenyl
CF_3	1-ethyl-2-butenyl
CF_3	1-isopropyl-2-butenyl
CF_3	1-n-butyl-2-butenyl
CF_3	1-methyl-2-pentenyl
CF_3	1,4-dimethyl-2-pentenyl
CF_3	propargyl
CF_3	2-butynyl
CF_3	3-butynyl
CF_3	ethoxy
CF_3	propoxy
CF_3	1-methylethoxy

R^1	R
CF_3	n-butoxy
CF_3	1-methylpropoxy
CF_3	2-methylpropoxy
CF_3	1,1-dimethylethoxy
CF_3	n-pentyloxy
CF_3	n-hexyloxy
CF_3	2-ethylhexyloxy
CF_3	2-propenyloxy
CF_3	2-butenyloxy
CF_3	2-methyl-2-propenyloxy
CF_3	2-pentenyloxy
CF_3	3-pentenyloxy
CF_3	3-chloro-2-propenyloxy
CF_3	2,3-dichloro-2-propenyloxy
CF_3	2,3,3-trichloropropenyloxy
CF_3	2-propynyloxy
CF_3	2-butynyloxy
CF_3	3-butynyloxy
CF_3	1-methyl-2-propynyloxy
CF_3	cyclopropyl
CF_3	cyclobutyl
CF_3	cyclopentyl
CF_3	cyclohexyl
CF_3	2-cyclopentenyl
CF_3	1-cyclopentenyl
CF_3	2-cyclohexenyl
CF_3	1-cyclohexenyl
CF_3	cyclopentyloxy
CF_3	cyclohexyloxy
CF_3	2-cyclopentenyloxy
CF_3	2-cyclohexenyloxy
CF_3	phenyl
Cl	i-C_3H_7
Cl	n-C_3H_7
Cl	n-C_4H_9
Cl	sec.-C_4H_9
Cl	i-C_4H_9
Cl	tert.-C_4H_9

R^1	R
Cl	n-C_5H_{11}
Cl	sec.-C_5H_{11}
Cl	n-C_6H_{13}
Cl	n-C_7H_{15}
Cl	sec.-C_7H_{15}
Cl	1-methylvinyl
Cl	2-methylvinyl
Cl	allyl
Cl	2-methylvinyl
Cl	2-ethylallyl
Cl	1-methylallyl
Cl	1-ethylallyl
Cl	1-methyl-2-butenyl
Cl	1-ethyl-2-butenyl
Cl	1-isopropyl-2-butenyl
Cl	1-n-butyl-2-butenyl
Cl	methyl-2-pentenyl
Cl	1,4-dimethyl-2-pentenyl
Cl	propargyl
Cl	2-butynyl
Cl	3-butynyl
Cl	ethoxy
Cl	propoxy
Cl	1-methylethoxy
Cl	n-butoxy
Cl	1-methylpropoxy
Cl	2-methylpropoxy
Cl	1,1-dimethylethoxy
Cl	n-pentyloxy
Cl	n-hexyloxy
Cl	2-ethylhexyloxy
Cl	2-propenyloxy
Cl	2-butenyloxy
Cl	2-methyl-2-propenyloxy
Cl	2-pentenyloxy
Cl	3-pentenyloxy
Cl	3-chloro-2-propenyloxy
Cl	2,3-dichloro-2-propenyloxy

R^1	R
Cl	2,3,3-trichloropropenyloxy
Cl	2-propynyloxy
Cl	2-butynyloxy
Cl	3-butynyloxy
Cl	1-methyl-2-propynyloxy
Cl	cyclopropyl
Cl	cyclobutyl
Cl	cyclopentyl
Cl	cyclohexyl
Cl	2-cyclopentenyl
Cl	1-cyclopentenyl
Cl	2-cyclohexenyl
Cl	1-cyclohexenyl
Cl	cyclopentyloxy
Cl	cyclohexyloxy
Cl	2-cyclopentenyloxy
Cl	2-cyclohexenyloxy
Cl	phenyl

Table B



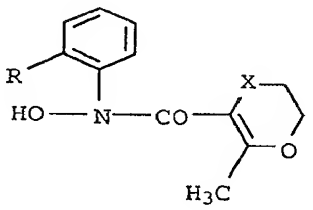
I.2

R^2	R
Cl	i-C_3H_7
Cl	n-C_3H_7
Cl	n-C_4H_9
Cl	sec.-C_4H_9
Cl	i-C_4H_9
Cl	tert.-C_4H_9
Cl	n-C_5H_{11}
Cl	sec.-C_5H_{11}
Cl	n-C_6H_{13}
Cl	n-C_7H_{15}
Cl	sec.-C_7H_{15}
Cl	1-methylvinyl
Cl	2-methylvinyl
Cl	allyl
Cl	2-methylallyl
Cl	2-ethylallyl
Cl	1-methylallyl
Cl	1-ethylallyl
Cl	1-methyl-2-butenyl
Cl	1-ethyl-2-butenyl
Cl	1-isopropyl-2-butenyl
Cl	1-n-butyl-2-butenyl
Cl	1-methyl-2-pentenyl
Cl	1,4-dimethyl-2-pentenyl
Cl	propargyl
Cl	2-butynyl
Cl	3-butynyl
Cl	ethoxy
Cl	propoxy
Cl	1-methylethoxy
Cl	n-butoxy

R^2	R
Cl	1-methylpropoxy
Cl	2-methylpropoxy
Cl	1,1-dimethylethoxy
Cl	n-pentyloxy
Cl	n-hexyloxy
Cl	2-ethylhexyloxy
Cl	2-propenyloxy
Cl	2-butenyloxy
Cl	2-methyl-2-propenyloxy
Cl	2-pentenyloxy
Cl	3-pentenyloxy
Cl	3-chloro-2-propenyloxy
Cl	2,3-dichloro-2-propenyloxy
Cl	2,3,3-trichloropropenyloxy
Cl	2-propynyloxy
Cl	2-butynyloxy
Cl	3-butynyloxy
Cl	1-methyl-2-propynyloxy
Cl	cyclopropyl
Cl	cyclobutyl
Cl	cyclopentyl
Cl	cyclohexyl
Cl	2-cyclopentenyl
Cl	1-cyclopentenyl
Cl	2-cyclohexenyl
Cl	1-cyclohexenyl
Cl	cyclopentyloxy
Cl	cyclohexyloxy
Cl	2-cyclopentenyloxy
Cl	2-cyclohexenyloxy
Cl	i-C_3H_7
Cl	n-C_3H_7
Cl	n-C_4H_9
Cl	sec.-C_4H_9
Cl	i-C_4H_9
Cl	tert.-C_4H_9
Cl	n-C_5H_{11}
Cl	sec.-C_5H_{11}

R^2	R
Cl	n-C_6H_{13}
Cl	n-C_7H_{15}
Cl	sec.-C_7H_{15}
Cl	ethoxy
Cl	propoxy
Cl	1-methylethoxy
Cl	n-butoxy
Cl	1-methylpropoxy
Cl	2-methylpropoxy
Cl	1,1-dimethylethoxy
Cl	n-pentyloxy
Cl	n-hexyloxy
Cl	cyclopentyl
Cl	phenyl

Table C



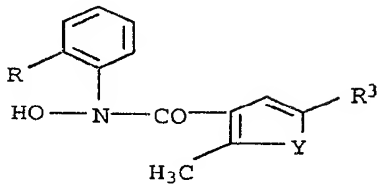
I.3

X	R
CH_2	i-C_3H_7
CH_2	n-C_3H_7
CH_2	n-C_4H_9
CH_2	sec.-C_4H_9
CH_2	i-C_4H_9
CH_2	tert.-C_4H_9
CH_2	n-C_5H_{11}
CH_2	sec.-C_5H_{11}
CH_2	n-C_6H_{13}
CH_2	n-C_7H_{15}
CH_2	sec.-C_7H_{15}
CH_2	1-methylvinyl
CH_2	2-methylvinyl
CH_2	allyl
CH_2	2-methylallyl
CH_2	2-ethylallyl
CH_2	1-methylallyl
CH_2	1-ethylallyl
CH_2	1-methyl-2-butenyl
CH_2	1-ethyl-2-butenyl
CH_2	1-isopropyl-2-butenyl
CH_2	1-n-butyl-2-butenyl
CH_2	1-methyl-2-pentenyl
CH_2	1,4-dimethyl-2-pentenyl
CH_2	propargyl
CH_2	2-butynyl
CH_2	3-butynyl
CH_2	ethoxy
CH_2	propoxy

X	R
CH_2	1-methylethoxy
CH_2	n-butoxy
CH_2	1-methylpropoxy
CH_2	2-methylpropoxy
CH_2	1,1-dimethylethoxy
CH_2	n-pentyloxy
CH_2	n-hexyloxy
CH_2	2-ethylhexyloxy
CH_2	2-propenyloxy
CH_2	2-butenyloxy
CH_2	2-methyl-2-propenyloxy
CH_2	2-pentenyloxy
CH_2	3-pentenyloxy
CH_2	3-chloro-2-propenyloxy
CH_2	2,3-dichloro-2-propenyloxy
CH_2	2,3,3-trichloropropenyloxy
CH_2	2-propynyloxy
CH_2	2-butynyloxy
CH_2	3-butynyloxy
CH_2	1-methyl-2-propynyloxy
CH_2	cyclopropyl
CH_2	cyclobutyl
CH_2	cyclopentyl
CH_2	cyclohexyl
CH_2	2-cyclopentenyl
CH_2	1-cyclopentenyl
CH_2	2-cyclohexenyl
CH_2	1-cyclohexenyl
CH_2	cyclopentyloxy
CH_2	cyclohexyloxy
CH_2	2-cyclopentenyloxy
CH_2	2-cyclohexenyloxy
S	i-C_3H_7
S	n-C_3H_7
S	n-C_4H_9
S	sec.-C_4H_9
S	i-C_4H_9
S	tert.-C_4H_9

X	R
S	n-C_5H_{11}
S	sec.-C_5H_{11}
S	n-C_6H_{13}
S	n-C_7H_{15}
S	sec.-C_7H_{15}
S	ethoxy
S	propoxy
S	1-methylethoxy
S	n-butoxy
S	1-methylpropoxy
S	2-methylpropoxy
S	1,1-dimethylethoxy
S	n-pentyloxy
S	n-hexyloxy
S	cyclopentyl
S	phenyl

Table D

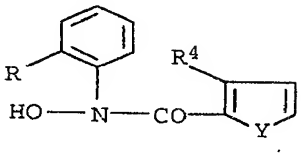


I.4

R^3	R	Y
H	i-C_3H_7	O
H	n-C_3H_7	O
H	n-C_4H_9	O
H	sec.-C_4H_9	O
H	i-C_4H_9	O
H	tert.-C_4H_9	O
H	n-C_5H_{11}	O
H	sec.-C_5H_{11}	O
H	n-C_6H_{13}	O
H	n-C_7H_{15}	O
H	sec.-C_7H_{15}	O
H	ethoxy	O
H	propoxy	O
H	1-methylethoxy	O
H	n-butoxy	O
H	1-methylpropoxy	O
H	2-methylpropoxy	O
H	1,1-dimethylethoxy	O
H	n-pentyloxy	O
H	n-hexyloxy	O
H	cyclopentyl	O
H	cyclohexyl	O
H	2-cyclopentenyl	O
H	1-cyclopentenyl	O
H	2-cyclohexenyl	O
H	1-cyclohexenyl	O
H	cyclopentyloxy	O
H	cyclohexyloxy	O
H	2-cyclopentenyloxy	O

R^3	R	Y
H	2-cyclohexenyloxy	O
CH_3	i-C_3H_7	O
CH_3	n-C_3H_7	O
CH_3	n-C_4H_9	O
CH_3	sec.-C_4H_9	O
CH_3	i-C_4H_9	O
CH_3	tert.-C_4H_9	O
CH_3	n-C_5H_{11}	O
CH_3	sec.-C_5H_{11}	O
CH_3	n-C_6H_{13}	O
CH_3	n-C_7H_{15}	O
CH_3	sec.-C_7H_{15}	O
CH_3	ethoxy	O
CH_3	propoxy	O
CH_3	1-methylethoxy	O
CH_3	n-butoxy	O
CH_3	1-methylpropoxy	O
CH_3	2-methylpropoxy	O
CH_3	1,1-dimethylethoxy	O
CH_3	n-pentyloxy	O
CH_3	n-hexyloxy	O
CH_3	cyclopentyl	O
CH_3	phenyl	O

Table E

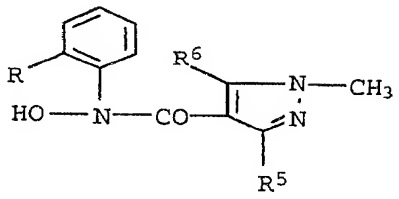


I.5

R^4	R	Y
CH_3	i-C_3H_7	O
CH_3	n-C_3H_7	O
CH_3	n-C_4H_9	O
CH_3	sec.-C_4H_9	O
CH_3	i-C_4H_9	O
CH_3	tert.-C_4H_9	O
CH_3	n-C_5H_{11}	O
CH_3	sec.-C_5H_{11}	O
CH_3	n-C_6H_{13}	O
CH_3	n-C_7H_{15}	O
CH_3	sec.-C_7H_{15}	O
CH_3	ethoxy	O
CH_3	propoxy	O
CH_3	1-methylethoxy	O
CH_3	n-butoxy	O
CH_3	1-methylpropoxy	O
CH_3	2-methylpropoxy	O
CH_3	1,1-dimethylethoxy	O
CH_3	n-pentyloxy	O
CH_3	n-hexyloxy	O
CH_3	cyclopentyl	O
CH_3	cyclopentenyl	O
CH_3	i-C_3H_7	S
CH_3	n-C_3H_7	S
CH_3	n-C_4H_9	S
CH_3	sec.-C_4H_9	S
CH_3	i-C_4H_9	S
CH_3	tert.-C_4H_9	S
CH_3	n-C_5H_{11}	S
CH_3	sec.-C_5H_{11}	S
CH_3	n-C_6H_{13}	S

R^4	R	Y
CH_3	n-C_7H_{15}	S
CH_3	sec.-C_7H_{15}	S
CH_3	ethoxy	S
CH_3	propoxy	S
CH_3	1-methylethoxy	S
CH_3	n-butoxy	S
CH_3	1-methylpropoxy	S
CH_3	2-methylpropoxy	S
CH_3	1,1-dimethylethoxy	S
CH_3	n-pentyloxy	S
CH_3	n-hexyloxy	S
CH_3	cyclopentyl	S
CH_3	cyclopentenyl	S

Table F



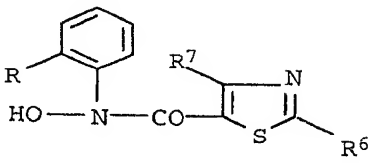
I.6

R^5	R^6	R
CH_3	H	i-C_3H_7
CH_3	H	n-C_3H_7
CH_3	H	n-C_4H_9
CH_3	H	sec.-C_4H_9
CH_3	H	i-C_4H_9
CH_3	H	tert.-C_4H_9
CH_3	H	n-C_5H_{11}
CH_3	H	sec.-C_5H_{11}
CH_3	H	n-C_6H_{13}
CH_3	H	n-C_7H_{15}
CH_3	H	sec.-C_7H_{15}
CH_3	H	1-methylvinyl
CH_3	H	2-methylvinyl
CH_3	H	allyl
CH_3	H	2-methylallyl
CH_3	H	2-ethylallyl
CH_3	H	1-methylallyl
CH_3	H	1-ethylallyl
CH_3	H	1-methyl-2-butenyl
CH_3	H	1-ethyl-2-butenyl
CH_3	H	1-isopropyl-2-butenyl
CH_3	H	1-n-butyl-2-butenyl
CH_3	H	1-methyl-2-pentenyl
CH_3	H	1,4-dimethyl-2-pentenyl
CH_3	H	propargyl
CH_3	H	2-butynyl
CH_3	H	3-butynyl
CH_3	H	ethoxy
CH_3	H	propoxy

R^5	R^6	R
CH_3	H	1-methylethoxy
CH_3	H	n-butoxy
CH_3	H	1-methylpropoxy
CH_3	H	2-methylpropoxy
CH_3	H	1,1-dimethylethoxy
CH_3	H	n-pentyloxy
CH_3	H	n-hexyloxy
CH_3	H	2-ethylhexyloxy
CH_3	H	2-propenyloxy
CH_3	H	2-butenyloxy
CH_3	H	2-methyl-2-propenyloxy
CH_3	H	2-pentenyloxy
CH_3	H	3-pentenyloxy
CH_3	H	3-chloro-2-propenyloxy
CH_3	H	2,3-dichloro-2-propenyloxy
CH_3	H	2,3,3-trichloropropenyloxy
CH_3	H	2-propynyloxy
CH_3	H	2-butynyloxy
CH_3	H	3-butynyloxy
CH_3	H	1-methyl-2-propynyloxy
CH_3	H	cyclopropyl
CH_3	H	cyclobutyl
CH_3	H	cyclopentyl
CH_3	H	cyclohexyl
CH_3	H	2-cyclopentenyl
CH_3	H	1-cyclopentenyl
CH_3	H	2-cyclohexenyl
CH_3	H	1-cyclohexenyl
CH_3	H	cyclopentyloxy
CH_3	H	cyclohexyloxy
CH_3	H	2-cyclopentenyloxy
CH_3	H	2-cyclohexenyloxy
CF_3	H	i-C_3H_7
CF_3	H	n-C_3H_7
CF_3	H	n-C_4H_9
CF_3	H	sec.-C_4H_9
CF_3	H	i-C_4H_9
CF_3	H	tert.-C_4H_9

R^5	R^6	R
CF_3	H	$n-C_5H_{11}$
CF_3	H	$sec.-C_5H_{11}$
CF_3	H	$n-C_6H_{13}$
CF_3	H	$n-C_7H_{15}$
CF_3	H	$sec.-C_7H_{15}$
CF_3	H	ethoxy
CF_3	H	propoxy
CF_3	H	1-methylethoxy
CF_3	H	n-butoxy
CF_3	H	1-methylpropoxy
CF_3	H	2-methylpropoxy
CF_3	H	1,1-dimethylethoxy
CF_3	H	n-pentyloxy
CF_3	H	n-hexyloxy
CF_3	H	cyclopentyl
CF_3	H	cyclopentenyl
CF_3	H	phenyl

Table G



I.7

R^7	R^6	R
CF_3	CH_3	i-C_3H_7
CF_3	CH_3	n-C_3H_7
CF_3	CH_3	n-C_4H_9
CF_3	CH_3	sec.-C_4H_9
CF_3	CH_3	i-C_4H_9
CF_3	CH_3	tert.-C_4H_9
CF_3	CH_3	n-C_5H_{11}
CF_3	CH_3	sec.-C_5H_{11}
CF_3	CH_3	n-C_6H_{13}
CF_3	CH_3	n-C_7H_{15}
CF_3	CH_3	sec.-C_7H_{15}
CF_3	CH_3	1-methylvinyl
CF_3	CH_3	2-methylvinyl
CF_3	CH_3	allyl
CF_3	CH_3	2-methylallyl
CF_3	CH_3	2-ethylallyl
CF_3	CH_3	1-methylallyl
CF_3	CH_3	1-ethylallyl
CF_3	CH_3	1-methyl-2-butenyl
CF_3	CH_3	1-ethyl-2-butenyl
CF_3	CH_3	1-isopropyl-2-butenyl
CF_3	CH_3	1-n-butyl-2-butenyl
CF_3	CH_3	1-methyl-2-pentenyl
CF_3	CH_3	1,4-dimethyl-2-pentenyl
CF_3	CH_3	propargyl
CF_3	CH_3	2-butynyl
CF_3	CH_3	3-butynyl
CF_3	CH_3	ethoxy
CF_3	CH_3	propoxy
CF_3	CH_3	1-methylethoxy

R^7	R^6	R
CF_3	CH_3	n-butoxy
CF_3	CH_3	1-methylpropoxy
CF_3	CH_3	2-methylpropoxy
CF_3	CH_3	1,1-dimethylethoxy
CF_3	CH_3	n-pentyloxy
CF_3	CH_3	n-hexyloxy
CF_3	CH_3	2-ethylhexyloxy
CF_3	CH_3	2-propenyloxy
CF_3	CH_3	2-butenyloxy
CF_3	CH_3	2-methyl-2-propenyloxy
CF_3	CH_3	2-pentenyloxy
CF_3	CH_3	3-pentenyloxy
CF_3	CH_3	3-chloro-2-propenyloxy
CF_3	CH_3	2,3-dichloro-2-propenyloxy
CF_3	CH_3	2,3,3-trichloropropenyloxy
CF_3	CH_3	2-propynyloxy
CF_3	CH_3	2-butynyloxy
CF_3	CH_3	3-butynyloxy
CF_3	CH_3	1-methyl-2-propynyloxy
CF_3	CH_3	cyclopropyl
CF_3	CH_3	cyclobutyl
CF_3	CH_3	cyclopentyl
CF_3	CH_3	cyclohexyl
CF_3	CH_3	2-cyclopentenyl
CF_3	CH_3	1-cyclopentenyl
CF_3	CH_3	2-cyclohexenyl
CF_3	CH_3	1-cyclohexenyl
CF_3	CH_3	cyclopentyloxy
CF_3	CH_3	cyclohexyloxy
CF_3	CH_3	2-cyclopentenyloxy
CF_3	CH_3	2-cyclohexenyloxy
CH_3	CH_3	i-C_3H_7
CH_3	CH_3	n-C_3H_7
CH_3	CH_3	n-C_4H_9
CH_3	CH_3	sec.-C_4H_9
CH_3	CH_3	i-C_4H_9
CH_3	CH_3	tert.-C_4H_9
CH_3	CH_3	n-C_5H_{11}

R^7	R^6	R
CH_3	CH_3	sec.-C_5H_{11}
CH_3	CH_3	n-C_6H_{13}
CH_3	CH_3	n-C_7H_{15}
CH_3	CH_3	sec.-C_7H_{15}
CH_3	CH_3	ethoxy
CH_3	CH_3	propoxy
CH_3	CH_3	1-methylethoxy
CH_3	CH_3	n-butoxy
CH_3	CH_3	1-methylpropoxy
CH_3	CH_3	2-methylpropoxy
CH_3	CH_3	1,1-dimethylethoxy
CH_3	CH_3	n-pentyloxy
CH_3	CH_3	n-hexyloxy
CH_3	CH_3	cyclopentyl
CH_3	CH_3	cyclopentenyl
CH_3	CH_3	phenyl

The novel active ingredients are particularly suitable for protecting various materials against degradation or destruction by bacteria or fungi or from being attacked by and covered with microorganisms. Examples of materials which can be preserved or microbicidally finished with the novel active ingredients are glues and adhesives, starch solutions, wax emulsions, clay emulsions, sizes, finishes, spinning baths, gelatine formulations, putty, joint sealants, cooling lubricants, drilling oils, fuels, plastic dispersions, emulsion paints, textiles, leather, raw hides and cosmetics. The compounds are also suitable as anti-slime agents in the paper industry, in cooling towers and in air moistening units.

The compounds I are also suitable for protecting the following plant species against attack by microorganisms:

cereals (e.g., wheat, barley, rye, oats, rice, sorghum and related species); beets (e.g., sugar and fodder beets); pomes, drupes and aggregate fruit (e.g., apples, pears, plums, peaches, almonds, cherries, strawberries, raspberries and blackberries); legumes (e.g., beans, lentils, peas, soybeans); oil-yielding crops (e.g., rape, mustard, poppies, olives, sunflowers, coconuts, castor-oil beans, cocoa beans, groundnuts); cucurbits (e.g., pumpkins, cucumbers, melons); fiber-yielding plants (e.g., cotton, flax, hemp, jute); citrus fruit (e.g., oranges, lemons, grapefruit, tangerines); vegetables (e.g., spinach, lettuce, asparagus, cabbage varieties, carrots, onions, tomatoes, potatoes, paprika); laurel species (e.g., avocado, cinnamomum, camphor) or plants such as Indian corn, tobacco, nuts, coffee, sugar cane, tea, grapes, hops, and banana and rubber trees. For the purposes of the present invention, the term "plants" is also taken to mean all types of other green growth, whether ornamentals, grassy areas, embankments, or generally low-growing cover crops.

For example the following microorganisms may be combatted with the novel compounds I:

Straphylococcus aureus, Escherichia coli, Klebsielle pneumoniae, Citrobacter freundii, Proteus vulgaris, Pseudomonas aeruginosa, Desulfovibrio desulfuricans, Streptoverticillium rubrireticuli, Aspergillus niger, Aspergillus versicolor, Penicillium funiculosum, Penicillium expansum, Penicillium glaucum, Paecilomyces variotii, Trichoderma viride, Chaetomium globosum, Aspergillus amstelodami, Phoma pigmentovora, Phoma violacea, Aureobasidium pullulans, Saccharomyces cerevisiae, Alternaria tenuis, Stemphylium macrosporoideum, Cladosporium herbarum, Cladosporium resinae, Candida albicans, Trichophyton mentagrophytes, Geotrichum candi-

dans, Monilia sitophila, Scenedesmus quadricauda, Chlorella vulgaris, Nostoc muscorium, Oscillatoria limosa and Anabaena constricta.

The novel substances can be converted into conventional formulations such as solutions, emulsions, suspensions, dusts, powders, pastes and granules. The application forms depend entirely on the purposes for which they are intended; they should at all events ensure a fine and uniform distribution of the active ingredient. The formulations are produced in known manner, for example by extending the active ingredient with solvents and/or carriers, with or without the use of emulsifiers and dispersants; if water is used as solvent, it is also possible to employ other organic solvents as auxiliary solvents. Suitable auxiliaries for this purpose are solvents such as aromatics (e.g., xylene), chlorinated aromatics (e.g., chlorobenzenes), paraffins (e.g., crude oil fractions), alcohols (e.g., methanol, butanol), ketones (e.g., cyclohexanone), amines (e.g., ethanolamine, dimethylformamide), and water; carriers such as ground natural minerals (e.g., kaolins, aluminas, talc and chalk) and ground synthetic minerals (e.g., highly disperse silica and silicates); emulsifiers such as nonionic and anionic emulsifiers (e.g., polyoxyethylene fatty alcohol ethers, alkyl sulfonates and aryl sulfonates); and dispersants such as lignin-sulfite waste liquors and methylcellulose.

The fungicides generally contain from 0.1 to 95, and preferably from 0.5 to 90, wt% of active ingredient. The active ingredients are used in a purity of from 90 to 100, and preferably from 95 to 100, % (according to the NMR/HPLC/GC spectrum).

Usual application concentrations are - based on the weight of the material to be protected - from 0.001 to 5, and preferably from 0.01 to 2, wt% of active ingredient; when the active ingredients are used for treating water, in oil production, in drilling and cutting oils, fuels, in swimming baths, cooling towers, air moistening units or in the paper industry, amounts of from 5 to 500 ppm are sufficient. Ready-to-use disinfectant solutions contain for instance from 0.5 to 10wt% of active ingredient.

Examples of such formulations are given below:

I. A solution of 90 parts by weight of compound no. 3 and 10 parts by weight of N-methyl-α-pyrrolidone, which is suitable for application in the form of very fine drops.

II. A mixture of 20 parts by weight of compound no. 4, 80 parts by weight of xylene, 10 parts by weight of the adduct of 8 to 10 moles of ethylene oxide and 1 mole of oleic acid-N-monoethanolamide, 5 parts by weight of the calcium salt of dodecylbenzenesulfonic acid, and 5 parts by weight of the adduct of 40 moles of ethylene oxide and 1 mole of castor oil. By finely dispersing the mixture in 100,000 parts by weight of water, an aqueous dispersion is obtained.

III. An aqueous dispersion of 20 parts by weight of compound no. 1, 40 parts by weight of cyclohexanone, 30 parts by weight of isobutanol, 20 parts by weight of the adduct of 40 moles of ethylene oxide and 1 mole of castor oil. A mixture of this dispersion with 100,000 parts by weight of water contains 0.02wt% of the active ingredient.

IV. An aqueous dispersion of 20 parts by weight of compound no. 3, 25 parts by weight of cyclohexanol, 65 parts by weight of a mineral oil fraction having a boiling point between 210 and 280°C, and 10 parts by weight of the adduct of 40 moles of ethylene oxide and 1 mole of castor oil. The mixture of this dispersion with 100,000 parts by weight of water contains 0.02wt% of the active ingredient.

V. A hammer-milled mixture of 80 parts by weight of compound no. 2, 3 parts by weight of the sodium salt of diisobutylnaphthalene-α-sulfonic acid, 10 parts by weight of the sodium salt of a lignin-sulfonic acid obtained from a sulfite waste liquor, and 7 parts by weight of powdered silica gel. By finely dispersing the mixture in 20,000 parts by weight of water, a spray liquor containing 0.1wt% of the active ingredient is obtained.

VI. An intimate mixture of 3 parts by weight of compound no. 1 and 97 parts by weight of particulate kaolin. The dust contains 3wt% of the active ingredient.

VII. An intimate mixture of 30 parts by weight of compound no. 4, 92 parts by weight of powdered silica gel and 8 parts by weight of paraffin oil sprayed onto the surface of this silica gel. This formulation of the active ingredient exhibits good adherence.

VIII. A stable aqueous dispersion of 40 parts by weight of compound no. 2, 10 parts of the sodium salt of a phenolsulfonic acid-urea-formaldehyde condensate, 2 parts of silica gel and 48 parts of water, which dispersion can be further diluted.

IX. A stable oily dispersion of 20 parts by weight of compound no. 3, 2 parts by weight of the calcium salt of dodecylbenzenesulfonic acid, 8 parts by weight of a fatty alcohol polyglycol ether, 2 parts by weight of the sodium salt of a phenolsulfonic acid-urea-formaldehyde condensate and 68 parts by weight of a paraffinic mineral oil.

X. A hammer-milled mixture of 10 parts by weight of compound no. 1, 4 parts by weight of the sodium salt of diisobutylnaphthalene-α-sulfonic acid, 20 parts by weight of the sodium salt of a lignin-sulfonic acid obtained from a sulfite waste liquor, 38 parts by weight of silica gel, and 38 parts by weight of kaolin. By finely dispersing the mixture in 10,000 parts by weight of water, a spray liquor containing 0.1wt% of the active ingredient is obtained.

Used alone, the active ingredients act as low-foaming biocides. A significant increase in the action of biocidal formulations containing these compounds is achieved if tri-C_6- to C_{12}-alkylmethylammonium salts, preferably in amounts of from 20 to 40wt%, based on the weight of compounds of the general formula I, are added.

The active ingredients may also be mixed with other, prior art, microbicides. In many instances, a synergistic effect is achieved, i.e., the microbicidal action of the mixture is greater than the added actions of its individual components.

Prior art microbicides may be added to the novel substances in a weight ratio of from 1:100 to 100:1.

Examples of such active ingredients are as follows:

2-(thiocyanomethylthio)-benzothiazole
1-[2-(2,4-dichlorophenyl)-2-(2-propenyloxy)-ethyl]-1H-imidazole
2,4,5,6-tetrachloroisophthalodinitrile
methylene bisthiocyanate
tributyltin oxide, naphthenate, benzoate, salicylate
mercaptobenzothiazole
1,2-benzisothiazolone and its alkali metal salts
alkali metal compounds of N'-hydroxy-N-cyclohexyldiazenium oxide
2-(methoxycarbonylamino)-benzimidazole
2-methyl-3-oxo-5-chlorothiazolin-3-one
trihydroxymethylnitromethane
glutardialdehyde
chloroacetamide
polyhexamethylene bisguanide
5-chloro-2-methyl-4-isothiazolin-3-one + magnesium salts

3,5-dimethyltetrahydro-1,3,5-2H-thiadiazine-2-thione
hexahydrotriazine
N,N-methylolchloroacetamide
2-n-octyl-4-isothiazolin-3-one
oxazolidines
bisoxazolidines
2,5-dihydro-2,5-dialkoxy-2,5-dialkylfurans
diethyldodecylbenzylammonium chloride
dimethyloctadecyldimethylbenzylammonium chloride
dimethyldidecylammonium chloride
dimethyldidodecylammonium chloride
trimethyltetradecylammonium chloride
benzyldimethylalkyl-(C_{12}-C_{18})-ammonium chloride
dichlorobenzyldimethyldodecylammonium chloride
cetylpyridinium chloride
cetylpyridinium bromide
cetyltrimethylammonium chloride
laurylpyridinium chloride
laurylpyridinium bisulfate
benzyldodecyldi(beta-oxyethyl)-ammonium chloride
dodecylbenzyltrimethylammonium chloride
n-alkyldimethylbenzylammonium chloride
(alkyl radical: 40% C_{12}, 50% C_{14}, 10% C_{16})
lauryldimethylethylammonium ethyl sulfate
n-alkyldimethyl-(1-naphthylmethyl)-ammonium chloride
(alkyl radical: 98% C_{12}, 2% C_{14})
cetyldimethylbenzylammonium chloride
lauryldimethylbenzylammonium chloride

Examples of further compounds which may be admixed are:

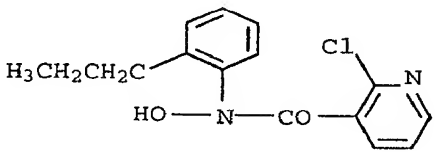
1,3-dimethylol-5,5-dimethylhydantoin
dimethylolurea
tetramethylolacetylenediurea
dimethylolglyoxalmonoureine
hexamethylenetetramine
glyoxal
glutardialdehyde
N-methylolchloroacetamide
1-(hydroxymethyl)-5,5-dimethylhydantoin
1,3-bis-(hydroxymethyl)-5,5-dimethylhydantoin
imidazolidinylurea
1-(3-chloroallyl)-3,5,7-triaza-1-azonia-adamantan chloride
1,3-bis-(β-ethylhexyl)-5-methyl-5-amino-hexahydropyrimidine
1,3,5-tris-(hydroxyethyl)-1,3,5-hexahydrotriazine
1,2-dibromo-2,4-dicyanobutane
5-bromo-5-nitro-1,3-dioxane

2-bromo-2-nitropropanediol
1,1'-hexamethylene-bis-[5-(4-chlorophenyl)-biguanide]
4,4-diaminodiphenoxypropane
2-bromo-2-nitropropane-1,3-diol
sorbic acid and its salts
p-hydroxybenzoic acid and its esters and salts
zinc-2-pyridinethiol-N-oxide
2-[(hydroxylmethyl)amino]-ethanol
dithio-2,2'-bis(benzmethylamide)
5-chloro-2-(2,4-dichlorophenoxy)-phenol
thio-bis-(4-chlorophenol)
o-phenylphenol
chloromethyl-diiodomethylsulfone
p-chlorophenyl-3-iodopropargylformal.

Synthesis examples

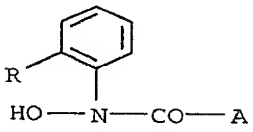
The directions given in the synthesis examples below were used, after appropriate modification of the starting materials, to obtain further compounds I. The compounds thus obtained are listed in the tables below with their physical data.

1. N-hydroxy-N-(2-propylphenyl)-2-chloronicotinamide



At 0°C, 14 ml of water and 19.6 g of sodium bicarbonate are added to a solution of 15.1 g of 2-n-propylphenylhydroxylamine in 75 ml of a 2:1 mixture of ether and ligroin, and 13.6 g of 2-chloronicotinamide is then dripped in while stirring vigorously. The mixture is stirred overnight at room temperature and then suction filtered. The residue is stirred for 15 minutes in 10% strength sodium bicarbonate solution, suction filtered, dissolved in ethyl acetate and dried, and the solvent is evaporated off under reduced pressure. From the crude product (14.6 g) there is isolated, after recrystallization from ethanol, 12.5 g of 2-chloronicotic acid-N-hydroxy-2-n-propylanilide of m.p. 134-135°C.

Table 1



I

Example no.	R	A	Phys. data
1	$CH(CH_3)_2$	2-Cl-pyridin-3-yl	107-111°C
2	$CH_2CH_2CH_3$	2-Cl-pyridin-3-yl	134-135°C
3	$CH_2CH(CH_3)_2$	2-Cl-pyridin-3-yl	oil
4	phenyl	2-Cl-pyridin-3-yl	112-115°C
5	$CH_2CH(CH_3)_2$	2-CH_3, 4-CF_3-thiazol-4-yl	oil
6	phenyl	2-CH_3, 4-CF_3-thiazol-4-yl	173-175°C
7	$CH_2CH(CH_3)_2$	2,4-$(CH_3)_2$-thiazol-4-yl	oil
8	phenyl	2,4-$(CH_3)_2$-thiazol-4-yl	58-62°C

Examples demonstrating biological action:

Action on Botrytis cinerea

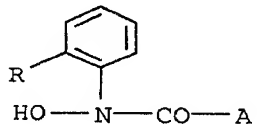
Slices of green paprika pods were sprayed to runoff with aqueous suspensions containing (dry basis) 80% of the active ingredient and 20% of emulsifier. After the sprayed-on layer had dried, the slices were sprayed with a spore suspension [$1.7 \cdot 10^6$ spores per ml; 2% biomalt; water] of the fungus Botrytis cinerea and then kept for 4 days at 18°C and in high humidity.

After this period, the untreated controls exhibited 90% fungus attack, whereas the paprika slices treated with 500 ppm of compounds nos. 1 and 2 exhibited 5% attack at most.

At an application rate of 1000 ppm of compounds nos. 1 and 2 the paprika slices exhibited no attack at all, whereas the slices treated with 1000 ppm of 2-chloronicotinic acid-2-chloroanilide exhibited 90% attack, just as the untreated controls.

We claim:

1. N-Hydroxy-N-phenylcarboxamides of the formula I



I

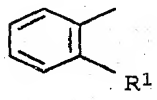
where:

R is C_2-C_{12}-alkyl, C_2-C_{12}-alkoxy, C_3-C_{12}-alkenyl, C_3-C_{12}-alkenyloxy, C_3-C_6-alkynyl or C_3-C_6-alkynyloxy, where these groups are partially or completely halogenated;

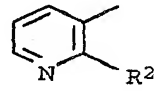
C_3-C_7-cycloalkyl, C_4-C_7-cycloalkenyl, C_3-C_7-cycloalkyloxy or C_4-C_7-cycloalkenyloxy, where these rings may bear from one to three C_1-C_4-alkyl groups;

phenyl, which may bear from one to five halogen atoms and/or from one to three of the following radicals: C_1-C_4-alkyl, C_1-C_4-haloalkyl, C_1-C_4-alkoxy, C_1-C_4-haloalkoxy, C_1-C_4-alkylthio or C_1-C_4-haloalkylthio;

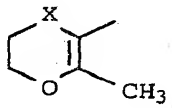
A is a cyclic radical selected from the group of formulae A1 to A7



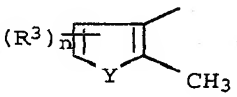
A1



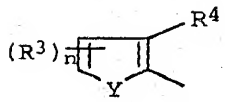
A2



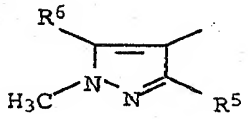
A3



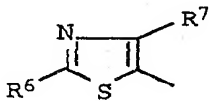
A4



A5



A6



A7

where:

X is $-CH_2-$, $-S-$, $-SO-$ or $-SO_2-$;
Y is $-O-$ or $-S-$;
R^1, R^2, R^4, R^5 and R^7 are halogen, C_1-C_4-alkyl or C_1-C_4-haloalkyl;
R^3 and R^6 are hydrogen, halogen or C_1-C_4-alkyl;
n is 1 or 2, and the radicals R^3 may be different when n is 2.

2. N-Hydroxy-N-phenylcarboxamides of the formula I as claimed in claim 1, where R has the meanings given in claim 1 and A is a cyclic radical selected from the group having the formulae A1 to A7, where X and Y have the meanings given in claim 1 and the substituents have the following meanings:

R^1 is halogen, methyl or C_1-haloalkyl;
R^2 is halogen or C_1-haloalkyl;
R^3 is hydrogen or methyl;
n is 1 or 2, and the radicals R^3 may be different when n is 2;
R^4 is halogen or methyl;
R^5 is methyl or C_1-haloalkyl;
R^6 is hydrogen, halogen or methyl;
R^7 is halogen, methyl or C_1-haloalkyl.

3. N-Hydroxy-N-phenylcarboxamides of the formula I as claimed in claim 1, where R has the meanings given in claim 1 and A is cyclic radical selected from the group having the formulae A1 to A7, where X and Y have the meanings given in claim 1 and the substituents have the following meanings:

R^1 is chloro, bromo, iodo, methyl or trifluoromethyl;
R^2 is chloro or trifluoromethyl;
R^3 is hydrogen or methyl;
n is 1 or 2, and the radicals R^3 may be different when n is 2;
R^4 is chloror or methyl;
R^5 is methyl, difluoromethyl or trifluoromethyl;
R^6 is hydrogen, chloro or methyl;
R^7 is chloro, methyl or trifluoromethyl.

4. An agent for combatting injurious fungi, containing a fungicidal amount of a compound of the formula I as claimed in claim 1, 2 or 3, and inert additives.

5. A process for combatting injurious fungi, wherein the fungi, their habitat and/or the plants or materials to be kept free from fungi are treated with a fungicidally effective amount of a compound of the formula I as claimed in claim 1, 2 or 3.

6. The use of compounds I as claimed in claim 1, 2 or 3 for combatting injurious fungi.

7. The use of compounds I as claimed in claim 1, 2 or 3 for combatting Botrytis.

SUBSTITUTE

REMPLACEMENT

SECTION is not Present

Cette Section est Absente